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Published in:
Physical Review B

Link to article, DOI:
10.1103/PhysRevB.49.12969

Publication date:
1994

Document Version
Publisher's PDF, also known as Version of record

Josephson properties of Nb/Cu multilayers

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(Received 27 August 1993; revised manuscript received 23 November 1993)

We report direct experimental observations of the Josephson properties of Nb/Cu multilayers achieved by measurements across layers. Multilayered sandwiches with a small cross section (20 μm in diameter) consisting of 10 Nb/Cu/Nb junctions in series were fabricated for such measurements. Applying RF power we observed Shapiro steps in the current-voltage characteristics. Changing the temperature we have seen the sequential change in the principal Shapiro-step voltage representing the change in the number of the phase-locked junctions. This phenomenon is direct evidence for the dimensional 3D-2D crossover responsible for the subdivision of the sample in the individual layers. Simultaneously with the change of the RF step voltage, the behavior of the I-V characteristics change. For $T < T_{2D}$ hysteresis appears in the I-V characteristics and the slope of the temperature dependence of the Josephson critical current, $I_c(T)$, changes.

I. INTRODUCTION

Superconducting multilayers reveal many interesting physical properties that are now intensively studied both from the point of view of their applications in superconducting electronics and in connection with high-temperature superconductors. Yet until now there is a lack of experimental results for multilayered structures, especially for measurements across layers. The main problem here is the fabrication of a sample consisting of a large number of stacked junctions with identical properties. Rather reproducible fabrication techniques has been achieved only for stacked double tunnel junction *SISIS* samples.[1-5] Physical properties of such double junction samples are very interesting and provide encouraging results from the point of view of their application in Josephson electronics. Among the applications of stacked junctions are the flux-flow oscillators[3] the Josephson voltage standard[4] and superconducting detectors.[6] Recently the successful fabrication of a sample with 10 Nb/Al-AlO$_x$/Nb stacked junctions was reported;[7] however, the properties of the junctions were not very close to each other. In the present paper we report the first successful fabrication of multilayered sandwiches for measurements across layers consisting of 10 *SNS* (Nb/Cu/Nb) stacked junctions with the properties very close to each other.

Although *SNS* multilayers have basic Josephson properties similar to *SIS* multilayers, they exhibit interesting properties inherent only in the *SNS* (*SS'S*) structures and caused by the proximity effect.[8-12] Typically the interlayer coupling in *SNS* multilayers is much larger than that of *SIS* multilayers. Even for *SIS* structures the finite electromagnetic coupling between layers strongly influences the properties of the multilayer. For example, the splitting of Swihart velocities in long stacked tunnel junctions was observed by Ustinov *et al.*[3] and in Refs. 2 and 7 very complicated Fraunhoffer patterns were observed for stacked tunnel junctions. Thus the effect of strong interlayer coupling should be even more pronounced for *SNS* multilayers.

One of the most important signatures of the strong coupling is the dimensional three-to-two-dimensional (3D-2D) crossover observed for *SNS* (*SS'S*) multilayers.[9,11-13] The dimensional 3D-2D crossover consists of the fact that at high temperatures, $T > T_{2D}$ (3D region), the multilayer behaves as a superconductor that is uniform across the layers. On the other hand, at low temperatures, $T < T_{2D}$ (2D region), the multilayer behaves as a stack of distinct layers. As shown in Ref. 12 the temperature T_{2D} could be significantly smaller than the critical temperature of the multilayer T_C if the transparency β of the interfaces (and thus the coupling between layers) is large enough. Quite naturally, the larger the coupling, the more uniform is the structure across layers. The 3D-2D crossover strongly influences the properties of multilayers.

Among the manifestations of the 3D-2D crossover we mention: (i) the change in the $H_{c2}^{\parallel}$ temperature dependence at $T = T_{2D}$.[9,11,13,14] At $T > T_{2D}$ we have $H_{c2}^{\parallel}(t) \sim (1 - T/T_c)$ as for the bulk superconductor (3D region) and at $T < T_{2D}$ we have $H_{c2}^{\parallel}(T) \sim (1 - T/T^*)^{1/2}$, $T^* \sim T_{2D}$, which is typical for a thin film in a parallel magnetic field (2D region). (ii) Recently evidence for the 3D-2D crossover was observed in the H_{c1} temperature dependence of the Nb/Cu multilayers.[9] In that case a sharp increase of the anisotropy of the lower critical field, $H_{c1}^{\perp}/H_{c1}^{\parallel}$, was obtained at $T<T_{2D}$ caused by a decrease of the core energy of the vortex parallel to layers in the 2D region when the core could be imbedded in N layers. (iii) A strong influence of the crossover on the temperature dependence of the critical current across layers I_c was observed in Ref. 12. Thus at $T<T_{2D}$ hysteresis in the current-voltage characteristics (IVC) appears, caused by a sharp increase of the effective junction capacitance in the 2D state. Simultaneously the slope of $I_c(T)$ change. In the 3D region, $T>T_{2D}$, $I_c \sim (1-T/T_c^{S/N})$ and in the 2D state $I_c \sim (1-T/T')$, with T' close to the critical temperature of the isolated Nb film. Qualitatively the 3D-2D crossover occurs when the coherence length across layers becomes of the order of the multilayer period $\xi_{\perp} \sim d$.[15] Since $\xi_{\perp}$ has the same temperature dependence as ξ_S, $\xi_{\perp} \sim (1-T/T_c)^{-1}$, it is increasing with increasing temperature. Thus a multilayer transits from the 3D to the 2D state with decreasing temperature.

Moreover a new type of a dimensional crossover could occur at low temperatures when the coherence length of N layers, $\xi_N(T)=(\hbar D_N/2\pi k_B T)^{1/2}$, increases so that the N-layer effective coherence length across layers $\xi_N^{\perp}$ becomes of the order of the N-layer thickness $\xi_N^{\perp} \sim \beta \xi_N \sim d_N$, where β is the transparency of the Nb/Cu interface. This new type of crossover corresponds to the case when the effective thickness of N layers, $d_N/\xi_N^{\perp}$ becomes small and thus the coupling between S layers increases. ξ_N is increasing with decreasing temperature. Thus contrary to the 3D-2D crossover the S/N multilayer transits from the 2D state to the 2D strongly coupled (2DSC) state, when the temperature is decreased. The 2D-2DSC crossover is associated with a rapid growth of the order parameter in N layers[10] and causes the increase of the lower critical field at low temperatures.[9,10,16] The temperature of the 2D-2DSC crossover observed for our multilayers is about 2–3 K (Ref. 9) in reasonable agreement with the transparency value, $\beta=0.2-0.4$,[8] and the ξ_N value.[17,18] Some experimental evidences of the 2D-2DSC crossover were observed in Refs. 9 and 13.

In the present paper we have studied experimentally the I-V (current-voltage) characteristics of Nb/Cu multilayers consisting of 10 stacked Nb/Cu/Nb junctions. The thickness of the Nb layers was $d_S=200$ Å and the thicknesses of the Cu layers were $d_N=150$ Å (sample I) and $d_N=200$ Å (sample II). The fabrication of *SNS* multilayers is much simpler than *SIS* multilayers. This is due to the fact that the critical current density of *SNS* junctions is less sensitive to the fabrication procedure than for *SIS* tunnel junctions. Thus it was possible to fabricate multilayers where all ten *SNS* junctions had properties very similar to each other. Applying rf power at x-band frequencies we observed the sequential increasing of the principal Shapiro step voltage with decreasing temperature. This phenomenon is caused by the sequential increase of the number of phase-locked parts in the multilayer. At high temperatures $T \sim T_c$ the principal Shapiro step voltage was $V_1=(h/2e)\nu$, ν is the applied rf frequency. Thus the whole multilayer behaves as a single weak link corresponding to the pure 3D state. On the other hand, at low temperatures the principal Shapiro step voltage was $V_{10}=10(h/2e)\nu$ representing the synchronized behavior of all ten junctions and corresponding to the pure 2D state. Thus we directly observed the 3D-2D crossover for our multilayers.

II. EXPERIMENTAL

The multilayered sandwiches for the measurements across layers were fabricated on a single crystalline Si substrate. The sample layout is shown in Fig. 1. The sample consists of thick top and bottom Nb electrodes (1500 Å), an isolating SiO_2 layer with a thickness 4000 Å and the multilayered sandwich. The electrode width was 100 μm. The Nb/Cu multilayers were prepared by rf sputtering with a bias voltage; no additional substrate heating was used. The multilayer consists of 10 Cu layers with the thickness $d_N=150$ Å (sample I) and $d_N=200$ Å (sample II) and ten Nb layers with the thickness $d_S=200$ Å. The multilayered sandwich was 20 μm in diameter, made small to increase the sample resistance. The pattern was formed by means of photolithography and rf sputter etching in an argon plasma with a carbon film mask. This method makes it possible to avoid shorts between the layers. To make contact between the top electrode and the multilayer a window of 12 μm in diameter was made by a chemical etching of SiO_2. To avoid the tunnel resistance between the superlattice sample and the top electrode the top Nb layer was cleaned by plasma etching before sputtering the top electrode. The fabrication procedure gives reproducible results. The normal-state resistances of samples I and II were $R_{N1}=7.0$ mΩ and $R_{N2}=7.2$ mΩ, respectively. The transverse resistivities of the multilayers are rather large. To our opinion these large values are caused by the tunnel resistances of the Nb/Cu interfaces that are inversely proportional to the transparency β.[19] The critical current at 4.2 K was about 230 mA for sample I and about 110 mA for sample II. Applying rf power with frequency ν at low temperatures we observed the fundamental Shapiro step at a voltage $V_{10}=10(h/2e)\nu$ for both samples. This means that both samples really consist of ten *SNS* junctions in series, i.e., there were no shorts of layers. Moreover, the properties of the 10 junctions were close to each other. This could be judged firstly because the single critical current was observed in the IVC's for all ten junctions and secondly,

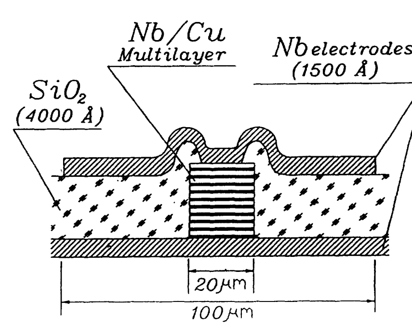

FIG. 1. Sketch of the multilayered Nb/Cu sample.

since it was possible to synchronize all junctions simultaneously by a very small rf power.

IVC's were obtained by a superconducting two-point measurement (no contact resistance) in the temperature range 4.2–9.0 K. For rf measurements the rf power from a sweep oscillator with a frequency range 8–18 GHz was supplied to the sample through a waveguide. The experimental setup had a copper shield to screen electromagnetic waves and a double μ metal can for shielding the magnetic field.

III. RESULTS AND DISCUSSION

A. Measurements without rf power

In Fig. 2 the IVC of sample I without rf power are shown for temperatures 7.1, 6.7, 6.3, 5.5, 4.9, and 4.3 K. Although the bias current is rather big, an estimate of the self-induced magnetic field shows that it is several times smaller than the lower critical field, $H_{c1}^{\parallel}(4.2\ \mathrm{K})\sim 10$ G.[9] From Fig. 2 it is seen that at low temperatures there is a pronounced hysteresis on the IVC, while at high temperatures it disappears. This hysteresis did not depend on the current sweep rate and amplitude and was not caused by sample heating. In Fig. 3 the critical currents I_c (open symbols) and the reverse currents I_R (stars) for both samples were shown. Here I_c is the current at which the sample switches from the superconducting to the resistive state when the current is increased and I_r is the current at which the sample returns to the superconducting state when the current is decreased. It is seen from Figs. 2 and 3 that a common feature for both samples is that at low temperatures there is a pronounced hysteresis in the IVC, while at high temperatures it disappears; note that the slopes of $I_c(T)$ change at the temperature where the hysteresis disappears. Moreover, the dashed lines in Fig. 3 show that at low temperature $I_c(T)$ for both samples

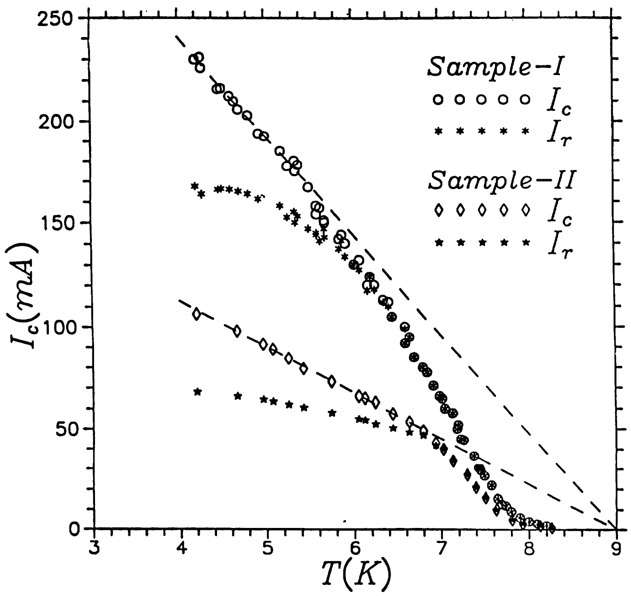

FIG. 3. Measured values of the critical current I_c and the reverse current I_r for samples I and II.

could be extrapolated to the temperature $T'=9$ K that is close to the critical temperature of the isolated Nb film. As it was shown in Ref. 12 all these phenomena are caused by the 3D-2D crossover. In particular, the disappearance of the hysteresis is caused by a significant decrease of the effective junction capacitance in the 3D state, when the sample becomes uniform across layers. The change of the temperature dependence of the critical current at low temperatures is caused by the fact that in the 2D region the order parameter of the S layers is close to that of the isolated superconducting film. The crossover temperature observed for Nb(200 Å)/Cu(150 Å) multilayers in Ref. 9, $T_{2D}=5.5$ K, is in good agreement with the observed temperature of the disappearance of the hysteresis for sample I.

The behavior of the sample II was similar to that of sample I. Yet, since the Cu layers were thicker in sample II, the critical current is smaller and the crossover temperature is larger.[12]

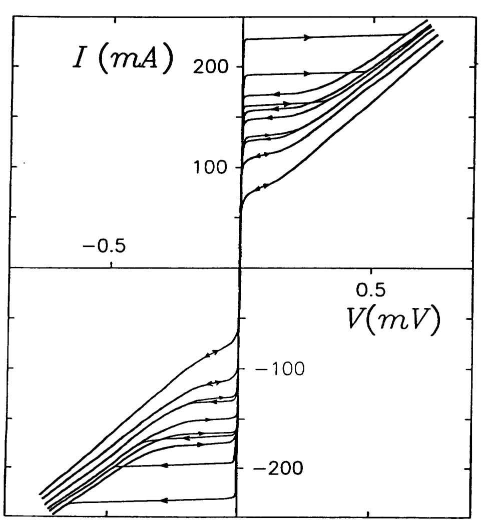

FIG. 2. The IVC of sample I without rf power for temperatures 7.1, 6.7, 6.3, 5.5, 4.9, and 4.3 K. It is seen that hysteresis of IVC appears at low temperatures.

B. Measurements with applied RF power

In Figs. 4 and 5 the IVC of sample I with applied rf power are shown for different temperatures. The rf power was small so that the suppression of the critical current was of the order of $\sim 10\%$. Figure 4 shows the I-V curves of sample I at high temperatures ($T=7.7$, 7.65, 7.6 K) and an applied frequency of $\nu=10.55$ GHz. To make the figure more clear the reverse branches (current decreasing) of IVC were shifted with respect to those of increasing current. Several voltage steps are seen in Fig. 4. The largest "principal" Shapiro step corresponds to the voltage, $V_1=21.8\ \mu\mathrm{V}\ \approx (h/2e)\nu$. Thus the multilayer is in the 3D state and behaves as a single junction. In accordance with the theory the sizes of higher-order steps are decreasing with number. In addition to the first and second Shapiro steps (see the arrow in Fig. 4) some subharmonic steps, $V_{n/m}=(h/2e(n/m)\nu$, were also observable. The appearance of these steps may be caused, e.g., by a nonsinusoidal current-phase relation $I_S(\varphi)$.[20] The reason for that could be the large distance between the Nb electrodes, $d_{\rm eff}\sim 4000$ Å. It is known that the $I_S(\varphi)$ relation becomes complicated when $d_{\rm eff}\gtrsim 3\xi_s$.[21] The fact that subharmonic steps decrease with increasing temperature (and consequently with increasing ξ_s) supports this assumption. The lower curve in Fig. 5 shows the IVC at $T=6.5$ K and $\nu=11.1$ GHz. This rf step corresponds to $N=4$, i.e., $V_4=91.8$ μV $\approx 4(h/2e)\nu$, indicating the existence of four synchronized junctions in series. The upper curve in Fig. 5 shows the IVC at $T=5.6$ K and $\nu=10.7$ GHz. The rf step voltage is $V_6=132.7$ μV $\approx 6(h/2e)\nu$, indicating the existence of the six synchronized junctions in series. In general, for sample I with the change of temperature it was possible to observe rf steps number 2, 4, 6, 8, and 10 (see Fig. 7, sample I), which are qualitatively similar to that in Fig. 5. The rf step at $V_{10}=10(h/2e)\nu$ corresponds to the pure 2D state when all ten junctions are distinct and synchronized. The voltage across each junction is $V_1=(h/2e)\nu$ but, since they are connected in series, the voltage on the multilayer is the sum of voltages of junctions.

The existence of only even steps for sample I shows that junction parameters of sample I are highly identical so that the sample divides in phase-locked parts symmetrically with respect to the center of the multilayer. For the sample II we also observed the increase of the principal Shapiro step voltage with a decrease of temperature. However, the behavior of sample II was more complicated. It was possible to observe also odd-numbered rf steps in the IVC (see Fig. 7, sample II), thus showing that the sample II was less perfect than sample I.

But the most striking feature of sample II was the existence of several rf steps at a time. In Fig. 6 the IVC for sample II with rf, $\nu=12.9$ GHz, at $T=4.3$ K is shown. It is seen that the sizes of rf steps with numbers 6, 7, 8, 9, and 10 are equal to each other, while all other steps are

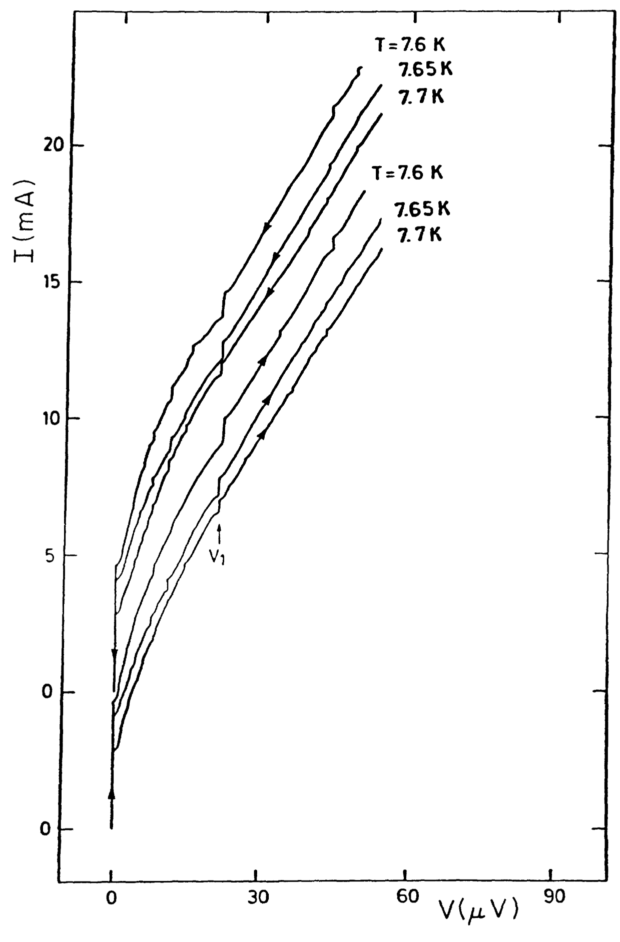

FIG. 4. The IVC of sample I at high temperatures ($T=7.7$, 7.65, 7.6 K) and an applied frequency of $\nu=10.55$ GHz. To make the figure more clear the reverse branches (current decreasing) of IVC were shifted with respect to those of increasing current. The arrow shows the principal Shapiro step at the voltage, $V_1=(h/2e)\nu\approx 21.8$ μV indicating that the multilayer is in the 3D state.

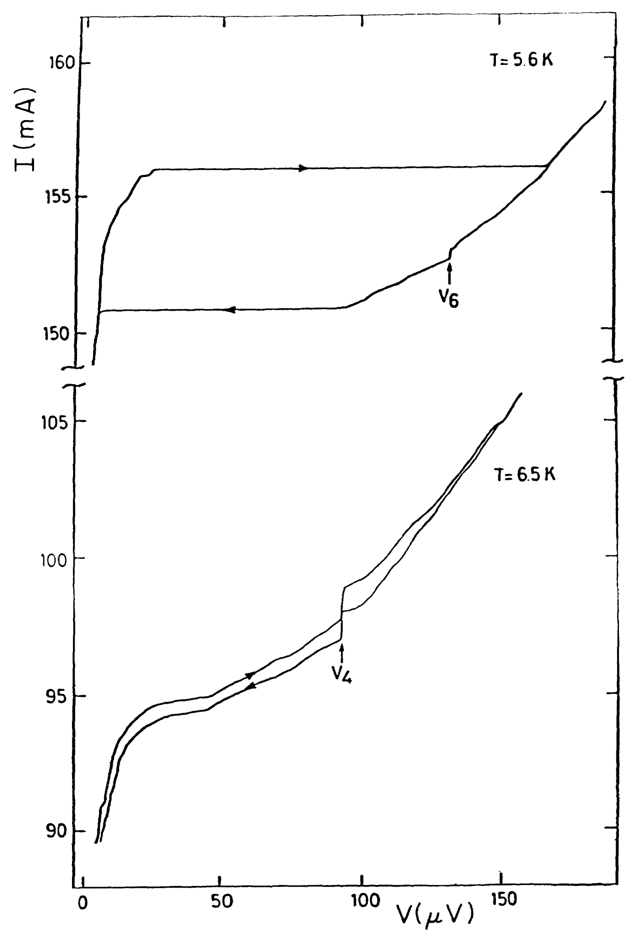

FIG. 5. The lower curve is the IVC of sample I at $T=6.5$ K and $\nu=11.1$ GHz. The rf step voltage is $V_4=4(h/2e)\nu\approx 91.8$ μV, indicating the existence of four synchronized junctions in series. The upper curve is the IVC of sample I at $T=5.6$ K and $\nu=10.7$ GHz. The rf step voltage is $V_6=6(h/2e)\nu\approx 132.7$ μV, indicating the existence of six synchronized junctions in series.

smaller. Since the rf power is very small, the size of step N ($N \neq 1$) should be smaller than that of the first step. Since the steps are vertical, they cannot be induced by synchronization of only a part of the junctions. Thus the data in Fig. 6 shows that for sample II the phase could be locked with different combinations of junctions. The fact that this phenomenon is more pronounced at low temperatures could be related with the increase of the coupling between layers near the second 2D-2DSC crossover when the temperature is decreased.[9,13] However, we cannot understand why this phenomenon occurs in the sample II and does not occur in the sample I. Presumably this is caused by the less perfect structure of the sample II, e.g., by the existence of superconducting microshorts that do not change significantly the critical current but can increase the coupling between layers.

Thus the picture of the physical processes taking place in the multilayer with the change of temperature is the following. Close to T_C the individual layers are not distinguishable and the sample behaves as a single S-S'-S (bottom Nb electrode–multilayer–top Nb electrode) junction. The multilayer is then in a pure 3D state. However, with the decrease of the temperature the multilayer is divided into phase-locked parts, and finally at low temperatures all ten junctions are distinct indicating the 2D state of the multilayer. The number of the principal Shapiro step as a function of the temperature for both samples is shown in Fig. 7. The increase of the number of phase-locked parts in the multilayers, giving the number of synchronized junctions with the decrease of temperature, is obvious.

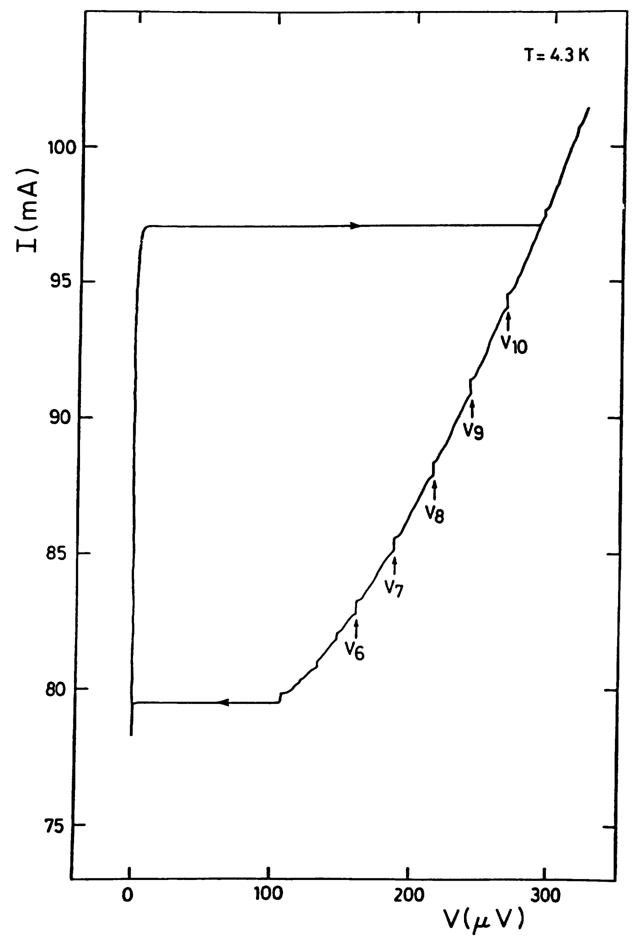

FIG. 6. The IVC of sample II at $T = 4.3$ K with rf frequency, $\nu = 12.9$ GHz. It is seen that the sizes of rf steps with numbers 6, 7, 8, 9, and 10 are equal to each other, while all other steps are smaller. Thus for sample II the phase could be locked with different combinations of junctions. The existence of the $N = 10$ step shows that the sample consists of 10 junctions in series.

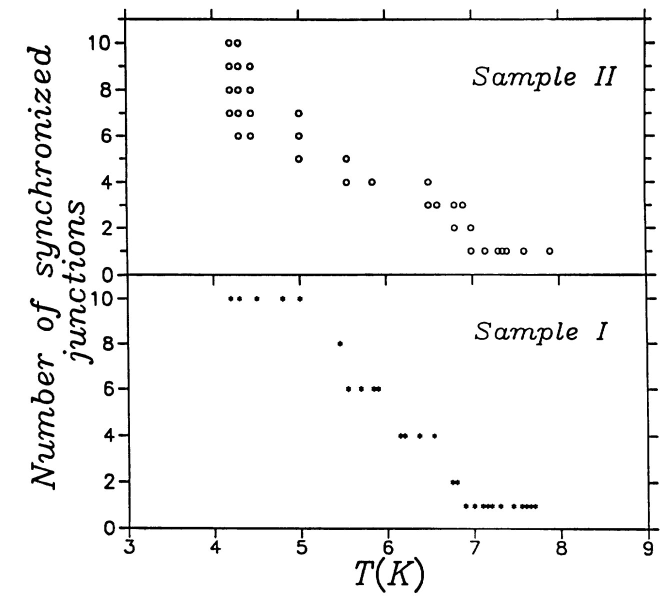

FIG. 7. The number of the principal Shapiro step as a function of the temperature for both samples. The increase of the number of phase-locked parts in the multilayers, giving the number of synchronized junctions with the decrease of temperature, is obvious. The case $N = 1$ at high temperatures corresponds to the pure 3D state of multilayers, while the case $N = 10$ at low temperatures represents the pure 2D state.

IV. CONCLUSIONS

In conclusion we have studied experimentally the Josephson properties and the current-voltage characteristics of the superconducting Nb (200 Å)/Cu(150 Å) and Nb(200 Å)/Cu(200 Å) multilayers for the current applied across layers. For this purpose unique samples consisting of ten stacked *SNS* junctions were fabricated. The strong influence of the dimensional 3D-2D crossover on the IVC was shown by the following observations: The hysteresis of the IVC diminishes at $T > T_{2D}$ and the slope of the temperature dependence of the critical current changes. Applying rf power we observed the synchronized behavior of all ten junctions at low temperatures. In general, it is not trivial to synchronize junctions in a series by a small rf power if their parameters differ slightly from each other. In our case the synchronization is achieved due to the strong coupling between junctions and due to the small spacing between layers. By decreasing the temperature we observed the process by which the sample is divided into phase-locked parts. This is the most direct observation of the dimensional 3D-2D crossover.

Superconducting multilayers are very promising objects for various practical applications such as the Josephson voltage standard and Josephson microelectronic devices. They could provide a significant increase of the integration level in Josephson microelectronics. Typically the useful properties of multilayers are related to synchronized behavior of the stacked junctions. The problem of synchronization is rather complicated, especially for junctions in series. One of the simplest methods to increase the mutual coupling of the junction is to decrease the thickness of the common electrode. This implies the fabrication of very thin layered structures. As we have shown in the current paper, in such structures the dimensional crossover could take place and one should take into account its dramatic influence on the physical properties of the structure.

ACKNOWLEDGMENTS

The authors are grateful to J. Mygind for helpful assistance in the experimental setup. The support from the NATO Linkage Grant No. LG 920672 is gratefully acknowledged.

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